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THE DISTRIBUTION OF THORIUM ISOTOPES BETWEEN LANTHANUM FLUORIDE CRYSTALS AND THEIR SATURATED SOLUTION IN 3 AND 12 PERCENT HNO_3 AT A TEMPERATURE OF 100 DEGREES

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The thorium isotope selected for this investigation was UX_1 and the system UX_1 (Th)F_4 - LaF_3, which is similar to the system YF_3 - CaF_2. Contrary to the view expressed by V. M. Goldschmidt, one of the authors, Khlopin (Zhurnal Fizicheskoy Khimii, Vol XIV, No 7, 1940), had previously assumed that systems of this type form pseudo or anomalous mixed crystals rather than genuine mixed crystals by penetration. If Goldschmidt's view is actually erroneous, a lower limiting proportion and a minimum concentration of one of the components must exist. Below this minimum concentration no mixed crystals are formed.

In view of the fact that the concentration of UX_1 in solution is extremely small -- i.e., of the order of $1 \cdot 10^{-12}$ M -- the lower limiting threshhold of concentration must have been passed. Under the circumstances mixed crystals could not have formed -- i.e., the precipitated crystals of LaF_3 did not contain any UX_1F_4; however, tables 1 and 2 of the original article show that this is not the case.

In order to study the distribution of UX_1 between the crystals and the solution of LaF_3, suspensions of LaF_3 in 3 and 12 percent nitric acid were prepared by precipitating the original nitric acid solutions (3 and 12 percent HNO_3) containing lanthanum nitrate with hydrofluoric acid. The precipitated lanthanum fluoride was washed with distilled water until the reactions for fluoride ions and nitrate ions became negative. Then saturated solutions of lanthanum fluoride in 3-percent nitric acid were prepared, adding enough lanthanum nitrate to offset the low solubility of the fluoride, so that the concentration in the solution could be brought up to 0.94 milligram lanthanum per milliliter of solution. The UX_1 was obtained in the usual manner from uranyl nitrate, except that precipitation was carried out on lanthanum hydroxide rather than iron. The hydroxide was then dissolved in 3 or 12 percent nitric acid and some of the resulting solution was added to the original solution of lanthanum nitrate and fluoride.

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Two suspensions of lanthanum fluoride were prepared, one inactive and the other active (containing UX_1). On proper combination of the active and inactive suspensions with active and inactive solutions respectively, mixtures containing definite amounts of lanthanum fluoride in the precipitated state and having the same volume were boiled under reflux for as long as 5 days in order to establish a state of equilibrium and determine the distribution of UX_1 between the active saturated lanthanum fluoride solution (in 3-percent HNO_3) and inactive LaF_3 crystals in one series (1) of experiments, and between active crystals and an identical inactive solution in another series (2). A constant acidity was maintained during the refluxing. The following coefficients of crystallization or distribution were found:

1.	(3% HNO_3)	K_{Φ}^{Φ} (D) = 1.68
2.	(3% HNO_3)	" = 1.75
1.	(12% HNO_3)	" = 1.35
2.	(12%)	" = 1.30

Furthermore, the rate at which equilibrium is established was determined in 3-percent HNO_3 at 100 degrees (i.e., by boiling). The results in question are summarized in Table 1, appended. They show that equilibrium is established beginning with the 48th hour, and that starting with the 8th hour the equilibrium value of K_{Φ}^{Φ} (D) can be determined with sufficient accuracy by taking the average of the two values listed in columns two and three of Table 1.

The results obtained in this investigation show that the distribution of UX_1 between the crystals of lanthanum fluoride and the latter salt's concentrated solution follows the law of Henry-Dalton-Berthelot-Nernst, i.e., that mixed crystals of UX_1F_4 and LaF_3 are actually formed. This can be reconciled with the authors' conception of the structure of these crystals and the assumption that they are anomalous mixed crystals only under one condition, namely, that a much higher concentration of thorium isotopes exists in the solution. It was acutally found that the lanthanum salts used in these experiments contained an admixture of thorium salts, a circumstance which resulted in a concentration of thorium isotopes close to 4.10^{-4} mM rather than one amounting to 1.10^{-12} mM. The thorium content in the lanthanum salts was determined by the radioactivity method according to the equivalent quantity of thoron.

In order to verify the conclusions in regard to the structure of the mixed crystals, experiments for the determination of the dependence of K_{Φ}^{Φ} (D) on the concentration of thorium isotopes in the concentration range of the latter from 10^{-2} to 10^{-7} mM were set up. The results, which have been listed in Table 2, clearly show that the value of K_{Φ}^{Φ} (D) approaches zero with diminishing concentrations. In other words, there is a lower limit of concentration.

Table 1. Formation of Equilibrium in the Distribution of UX_1 between Lanthanum Fluoride Crystals and the Saturated Solution of That Salt in 3-Percent HNO_3 at a Temperature of 100 Degrees

Time of Recrystallization (hr)	K_{Φ}^{Φ}(D) Active Solution -- Inactive Suspension	K_{Φ}^{Φ}(D) Inactive Solution -- Active Suspension
1	0.60	4.80
4	0.90	3.00
8	1.16	2.47
24	1.28	2.13
48	1.60	1.88
72	1.80	1.80
120	1.73	1.82

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Table 2. Dependence of $K_D(D)$ for UX_1 on the Total Concentration of Thorium Isotopes in Solution

Concentration of Thorium Isotopes in Solution (in mM)	Value of $K_D(D)$
1.2×10^{-2}	3.32
4.0×10^{-3}	2.77
2.0×10^{-3}	2.26
4.0×10^{-4}	1.71
2.0×10^{-5}	0.96
2.10×10^{-7}	0.17

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